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January 22, 1998

Dr. Alexander Pechenik  
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Dear Dr. Pechenik:

Enclosed please find the Final Technical Report for Grant No. F49620-96-1-0427, "Scale-up Equipment for Nano-size Particle Synthesis by Spray Pyrolysis."

Best regards,

Gary L. Messing

GLM/wb

cc: R. Killoren

## Final Technical Report

### "Scale-up Equipment for Nanosize Particle Synthesis by Spray Pyrolysis"

AFOSR Contract F49620-94-1-0427  
Dr. Alexander Pechenik, Project Monitor

We purchased a PSD52 Laboratory Spray Dryer for Closed Circuit Operation from APV Anhydro Separation Technologies. The equipment operates at 350°C using nitrogen gas with an outlet temperature of 100°C. Both aqueous and non-aqueous systems can be used in this system. This allows for organic solvent based systems to be used. Organic solvents can be recovered in an activated carbon assembly. For water drying, the drying capacity is 4 kg/h whereas acetone is 21 kg/h.

The system was delivered in November 1997 and will be installed in January 1998. However, it will be used for powder synthesis in the following DOD projects:

#### ARPA/AFOSR Supported Program

Grant #49620-94-1-0428, P.I.s G. L. Messing and S. Trolier-McKinstry  
Grant #DAAH04-95-1-0484, P.I. G. L. Messing

The goal of the ARPA/AFOSR supported research program is to establish a scientific foundation for the low cost preparation of bulk single crystal quality materials by templated grain growth (TGG); a process in which solid phase epitaxy is used to initiate solid state grain growth of a specific orientation in a polycrystalline matrix. We are investigating two classes of TGG; surface TGG in which a single crystal is contacted with a dense, polycrystalline ceramic and *in situ* TGG in which anisometric particles are uniformly dispersed and oriented in a polycrystalline matrix. The matrix must be fully dense and consist of sub-100 nm scale grains for TGG to work.  $\text{Bi}_4\text{Ti}_3\text{O}_{12}$  and  $\text{Sr}_2\text{Nb}_2\text{O}_7$  have been selected because of the need for high temperature piezoelectrics. These materials can be poled only when they are highly oriented materials.

We are also studying TGG for the low cost preparation of bulk single crystal quality  $\text{BaTiO}_3$  and  $(\text{Ba}_{1-x}\text{Sr}_x)\text{TiO}_3$ . This process is designed to provide large single crystals at considerably less expense than top-seeded solution grown  $\text{BaTiO}_3$  which cost on the order of \$5000 per crystal (Sanders Inc., Nashua, New Hampshire). Ultimately, such crystals are attractive for photorefractive applications including optical storage, phase conjugate mirrors, and multiple-wave mixing.

A key component of these projects is the availability of substantial quantities of high purity, nanosize powders with controlled stoichiometry. While nanosize  $\text{BaTiO}_3$  powder is commercially available it requires doping to achieve the desired dielectric properties. There is no commercial source for nanosize  $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ ,  $\text{Sr}_2\text{Nb}_2\text{O}_7$  and  $(\text{Ba}_{1-x}\text{Sr}_x)\text{TiO}_3$  powder. While we have developed processes for the synthesis of such powders, our SP processes are scale-limited. The ability to produce a kilogram of nanosize powder of these compositions and doped versions of these powders will have a profound effect on the project by allowing more effort to be spent on obtaining dense and submicrometer scale microstructures, grain growth studies and characterization. Also, a much larger range of compositions can be evaluated.

#### ARPA/ONR Supported Program

Grant #N00014-93-1-0502, P.I.s G. L. Messing and S. Trolier-McKinstry

The goal of this project is to produce oriented PMN-PT by *in situ* templated grain growth (TGG). Because of the high degree of texture possible by TGG we will be able to determine, for the first time, whether or not it is possible to achieve large strain actuators in polycrystalline ceramics.

PMN-PT is chosen as the primary material for investigation instead of PZN-PT due to the stability of the perovskite crystal structure. PZN-PT powder, on the other hand, when heated above  $\sim 500^{\circ}\text{C}$ , tends to decompose into a pyrochlore structure. Since the ceramic processing techniques enable excellent control of compositional homogeneity, preparation of oriented PMN-PT ceramics near the morphotropic phase boundary should be easier to produce than by single crystal growth techniques, where the high PT content is problematic in terms of uniformity.

Our overall goal is to produce a fine-grained (i.e.,  $<5\text{ }\mu\text{m}$ ), textured PMN-PT. Based on our earlier successes with textured  $\text{Sr}_2\text{Nb}_2\text{O}_7$  we know that we can refine the grain size by using a larger number of smaller seed particles to induce texture development. The acicular template particles seem to be particularly attractive for this purpose because they have diameters of  $<100\text{ nm}$  and aspect ratios of  $>10$ . To obtain fine-grained PMN-PT it may be necessary to use a high concentration of PT particles and thus, to achieve the target PMN-PT phase composition, will require compositional compensation of the matrix powder.

We have extensive experience at MRL in producing PMN and PMN-PT powders and ceramics. The formation of perovskite PMN is extremely sensitive to preparation conditions whereas PMN-PT can be produced by the columbite mixed oxide approach. Likewise, solution synthesis of perovskite PMN-PT is well documented. In this program, however, fine grain size is critical for both TGG and for the final component. Thus, we will use a chemical synthesis approach so we can sinter at a lower temperature and achieve fine grain size for templated grain growth.

The new spray pyrolysis system will allow further preparation of nanosize PMN, PMN-PT, PZN and PZT powders. Such fine powders are essential for the development of textured piezoelectrics by templated grain growth. This work will commence in March 1998.